

# Light quantization for arbitrary scattering systems

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We present a quantum theory of light scattering for the analysis of the quantum statistical and fluctuation properties of light scattered or emitted by micrometric and nanometric three-dimensional structures of arbitrary shape. We obtain general three-dimensional quantum-optical input-output relations providing the output photon operators in terms of the input photon operators and of the noise currents of the scattering system. These relations hold also for photon operators associated with evanescent fields, for anisotropic scattering systems and/or for media with a nonlocal susceptibility. We find that the commutation relations of the output photon operators, carrying all the information on the scattering and/or the emission process, result to be fixed by energy conservation and reciprocity. We prove that this quantization scheme is consistent with QED commutation rules by using a novel relationship between vacuum and thermal fluctuations. This theoretical framework has been applied to analyze the spectral density of light close to a point scatterer under different nonequilibrium conditions.

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## I. INTRODUCTION

The study of the optical properties of systems with shapes and sizes varying on micrometric and nanometric scales is motivated by fundamental research and by possible applications. In particular, the tailoring of electromagnetic modes allowed by microcavities and photonic bandgaps (PBGs) has given rise to a variety of striking phenomena observed in recent years [1,2] and it is expected to dramatically improve the performance of light-emitting devices. These developments and recent continuous progress in scanning near-field optical microscopy have stimulated new theoretical approaches for the analysis of a large class of problems dealing with three-dimensional (3D) objects of arbitrary shape and dielectric functions [3,4], and have renewed the interest in the classical theory of light scattering [5,6].

In this paper we present a quantum generalization of the classical theory of light scattering based on Green's dyadic technique. The theory presented here provides a general and unified basis for analyzing a large class of optical processes where quantum and/or thermal fluctuations play a role [7]. It is expected to be adequate to analyze a wide range of optical phenomena and experiments such as precision measurements of Casimir forces [8], light emission from sources embedded in photonic systems [9], light fluctuations in finite inverted-population media with inclusion of spatial effects [10], the spatial behavior of scattered and/or confined nonclassical light [11,12], nanoscale radiative transfer [13,14], etc.

Quantum electrodynamics in the presence of media started from the pioneering work by Agarwal [15] who, applying the fluctuation-dissipation theorem, developed the linear response theory of spontaneous emission in presence of dielectrics and conductors. A more direct microscopic quantization procedure for light in a dispersive and absorptive homogeneous dielectric was first proposed by Huttner and Barnett [16]. Since this work, following the method of Langevin forces, light has been quantized in media of increasing generality [17–23]. Within this method quite gen-

eral results have been recently presented [24,25]. In particular it has been proved that quantization of the Maxwell theory of the electromagnetic field in inhomogeneous three-dimensional, dispersive, and absorbing dielectric media of given causal permittivity is consistent with the fundamental equal-time commutation relations of QED [25]. A different general 3D quantization scheme that makes use of a set of auxiliary fields, followed by a canonical quantization procedure has been developed by Tip [26]. Recently the equivalence of the quantization schemes by Scheel *et al.* [25] and by Tip [26] has been demonstrated [27].

Here we generalize these results to media that can be anisotropic and/or with a nonlocal susceptibility. Furthermore, we consider explicitly media that can have finite size. This allows the analysis of quantized light scattering and allows us to derive general quantum-optical input-output relations relating the output photon operators to the input photon operators and to the noise currents of the scattering system. These relations hold also for evanescent fields and thus allow us to define naturally output photon operators associated with evanescent waves.

## II. THE SCATTERING SYSTEM PROPERTIES

Let us consider the most general nonmagnetic linear scattering system. It can be described by a causal and eventually nonlocal susceptibility tensor  $\chi_{i,j}(\mathbf{r}, \mathbf{r}', \omega)$  [28]. Thus we are considering a large class of material systems of arbitrary shape including anisotropic media and/or media driven by the electric field via a nonlocal susceptibility. Electronic states of semiconductors and semiconductor quantum structures, and also all those systems with a spatially dispersive susceptibility are driven by the electric field via a nonlocal susceptibility [21].

In the following we will adopt the compact Dirac notation. We introduce the operators  $\mathbf{L}$  for  $-\nabla \times \nabla \times$ ,  $\mathbf{e}_0$  for  $k^2 \mathbf{1}$ , and the integral operator  $\mathbf{e}_s$ , describing the effect of the scattering system. This operator applied to the electric field gives

$$\langle \mathbf{r}, i | \mathbf{e}_s | \mathbf{E} \rangle = k^2 \int d^3 r' \chi_{ij}(\mathbf{r}, \mathbf{r}') E_j(\mathbf{r}'). \quad \bar{N}(T) = \frac{N_u}{N_l - N_u}. \quad (2.7)$$

Hence the wave equation relative to the material systems here considered, for the positive frequency components of the electric-field operator can be written in the compact Dirac notation as

$$(\mathbf{L} + \mathbf{e}_0 + \mathbf{e}_s) | \hat{\mathbf{E}}^+ \rangle = i \omega \mu_0 | \hat{\mathbf{j}} \rangle, \quad (2.1)$$

where the hat indicates quantum operators. The zero mean noise currents  $\hat{\mathbf{j}}$  can be derived from the Heisenberg-Langevin equations for the material system [29] and appear only if the susceptibility tensor is not real; they are a direct consequence of the fluctuation-dissipation theorem and obey the following commutation rules,

$$[\hat{j}_i(\mathbf{r}, \omega), \hat{j}_j(\mathbf{r}', \omega)] = 0, \quad (2.2)$$

$$[\hat{j}_i(\mathbf{r}, \omega), \hat{j}_j^\dagger(\mathbf{r}', \omega)] = \frac{\hbar}{\pi \mu_0} \frac{\omega^2}{c^2} |\chi_{ij}^I(\mathbf{r}, \mathbf{r}', \omega)| \delta(\omega - \omega'), \quad (2.3)$$

$\chi^I$  being the imaginary part of the susceptibility tensor. These equations show that a nonlocal susceptibility produces noise currents that are spatially correlated. These spontaneous currents act as quantum Langevin forces. Their expectation values determine the amounts of noise that are added to optical signals that propagate through the attenuating or amplifying media. Moreover,  $\langle \hat{\mathbf{j}}^\dagger \hat{\mathbf{j}} \rangle$  is the source term producing light emission. These noise currents are related to the Bosonic vector field describing the reservoir oscillators. The expectation values of noise currents depend on the specific state of the reservoir oscillators. We start by considering a system at a given temperature  $T$ . In this case the current's correlation tensor is given by

$$\langle \hat{j}_i^\dagger(\mathbf{r}, \omega) \hat{j}_j(\mathbf{r}', \omega) \rangle = \frac{\hbar}{\pi \mu_0} \frac{\omega^2}{c^2} \chi_{ij}^I(\mathbf{r}, \mathbf{r}', \omega) \bar{N}(\omega, T) \delta(\omega - \omega'), \quad (2.4)$$

where  $\bar{N}(T)$  is the mode occupation described by Planck's formula

$$\bar{N}(\omega, T) = \frac{1}{\exp(\hbar \omega / k_B T) - 1}. \quad (2.5)$$

If we consider the medium composed of a collection of non-interacting two-level systems at thermal equilibrium, the ratio between the upper  $N_u$ - and the lower  $N_l$ -level occupations associated with the dielectric response at frequency  $\omega$  is given by the Boltzmann distribution law

$$\frac{N_u}{N_l} = \exp(-\hbar \omega / k_B T). \quad (2.6)$$

From Eqs. (2.5) and (2.6) we obtain

Thermal equilibrium can be altered by, e.g., optical pumping. In this case, Eq. (2.7) can still be used considering  $T \equiv T(\omega)$  as a frequency-dependent effective temperature. Very high temperatures correspond to saturation of the transition between the two levels. By taking a negative effective temperature, it is possible to describe also population inversion and hence amplifying media [30]. Equation (2.4) can also be generalized to take into account a scattering system including media with different effective temperatures. We obtain

$$\begin{aligned} \langle \hat{j}_i^\dagger(\mathbf{r}, \omega) \hat{j}_j(\mathbf{r}', \omega) \rangle &= \frac{\hbar}{\pi \mu_0} \frac{\omega^2}{c^2} \\ &\times \sum_m (\chi_m^I)_{ij}(\mathbf{r}, \mathbf{r}', \omega) \bar{N}(T_m, \omega) \\ &\times \delta(\omega - \omega'), \end{aligned} \quad (2.8)$$

where  $m$  labels the different media. We point out that the nonlocal susceptibility  $(\chi_m^I)_{ij}(\mathbf{r}, \mathbf{r}', \omega)$  is different from zero only if  $\mathbf{r}$  and  $\mathbf{r}'$  belong to the same medium  $m$ . The correlation  $\langle \hat{j}_i(\mathbf{r}, \omega) \hat{j}_j^\dagger(\mathbf{r}', \omega) \rangle$  can be obtained from Eq. (2.4) replacing  $\bar{N}$  with  $\bar{N} + 1$ .

### III. QUANTUM THEORY OF LIGHT SCATTERING

In the absence of the scattering system, the electric-field operator can be derived following the well-known quantization schemes in vacuum. By using the angular spectrum of plane waves, the electric-field operator can be expanded in terms of photon operators as

$$\hat{\mathbf{E}}^0(\mathbf{r}, t) = \int_0^\infty d\omega e^{-i\omega t} \hat{\mathbf{E}}^{0+}(\mathbf{r}, \omega) + \text{H.c.},$$

with

$$\hat{\mathbf{E}}^{0+}(\mathbf{r}, \omega) = i \sqrt{\frac{\hbar \omega}{2 \epsilon_0}} \sum_{\tau, K} \boldsymbol{\phi}_K^\tau(\mathbf{r}, \omega) \hat{a}_K^\tau(\omega), \quad (3.1)$$

where  $\tau = >, <$  indicates leftward and rightward propagating waves, and  $K \equiv (\mathbf{K}, \sigma)$  is a shortcut for the wave-vector projection along the  $xy$  plane and the polarization direction  $\sigma$ .  $\hat{a}_K^\tau$  are the photon operators obeying the usual Bosonic commutation rules,

$$[\hat{a}_K^\tau(\omega), \hat{a}_{K'}^{\tau'\dagger}(\omega')] = \delta_{\tau, \tau'} \delta_{K, K'} \delta(\omega - \omega'), \quad (3.2)$$

$$[\hat{a}_K^\tau(\omega), \hat{a}_{K'}^{\tau'}(\omega')] = 0. \quad (3.3)$$

The orthonormal set of vector fields is given by

$$\boldsymbol{\phi}_K^{> / <}(\mathbf{r}, \omega) = \alpha_{\mathbf{K}} \mathbf{e}_K^{> / <} \exp i(\mathbf{K} \cdot \mathbf{R} \pm k_z z), \quad (3.4)$$

where  $\mathbf{r}=(\mathbf{R},z)$ ,  $\mathbf{e}_k^\tau$  is the polarization unit vector,  $k_z=(\omega^2/c^2-K^2)^{1/2}$ , and  $\alpha_{\mathbf{K}}=(\omega/2\pi c^2 k_z \mathcal{A})^{1/2}$ ,  $\mathcal{A}$  being the quantization surface. In compact notation, Eq. (3.1) can be written as

$$|\hat{\mathbf{E}}^{0+}\rangle=i\sqrt{\frac{\hbar\omega}{2\varepsilon_0\tau,K}}\sum_{\tau,K}|\phi_k^\tau\rangle\hat{a}_K^\tau(\omega). \quad (3.5)$$

We observe that the field operator in Eq. (3.1) verifies, in the same compact notation of Eq. (2.1), the following wave equation:

$$(\mathbf{L}+\mathbf{e}_0)|\hat{\mathbf{E}}^{0+}\rangle=0. \quad (3.6)$$

The Green operator associated with the complete system is defined by

$$(\mathbf{L}+\mathbf{e}_0+\mathbf{e}_s)\mathbf{G}=\mathbf{1}. \quad (3.7)$$

Adding Eq. (3.6) to Eq. (2.1) and using Eq. (3.7) we obtain

$$|\hat{\mathbf{E}}^+\rangle=|\hat{\mathbf{E}}_h^+\rangle+|\hat{\mathbf{E}}_p^+\rangle, \quad (3.8)$$

with the particular solution  $\hat{\mathbf{E}}_p^+$  given by

$$|\hat{\mathbf{E}}_p^+\rangle=i\omega\mu_0\mathbf{G}|\hat{\mathbf{j}}\rangle, \quad (3.9)$$

and the homogeneous solution

$$|\hat{\mathbf{E}}_h^+\rangle=(\mathbf{1}-\mathbf{G}\mathbf{e}_s)|\hat{\mathbf{E}}^{0+}\rangle. \quad (3.10)$$

In the  $\mathbf{r}$  representation the obtained electric-field operator can be written as

$$\hat{E}_{hi}^+(\mathbf{r})=\hat{E}_i^{0+}(\mathbf{r})-q^2\int G_{ij}(\mathbf{r},\mathbf{r}')\chi_{jl}(\mathbf{r}',\mathbf{r}'')\hat{E}_l^{0+}(\mathbf{r}'')d\mathbf{r}'d\mathbf{r}'', \quad (3.11)$$

$$\hat{E}_{pi}^+(\mathbf{r})=i\omega\mu_0\int G_{ij}(\mathbf{r},\mathbf{r}')\hat{j}_j(\mathbf{r}')d\mathbf{r}'. \quad (3.12)$$

Introducing Eq. (3.1) into Eq. (3.10), the homogeneous term can be expanded in terms of free-space photon operators as

$$|\hat{\mathbf{E}}^+\rangle=i\sqrt{\frac{\hbar\omega}{2\varepsilon_0\tau,K}}\sum_{\tau,K}|\psi_k^\tau\rangle\hat{a}_K^\tau(\omega), \quad (3.13)$$

where

$$|\psi_k^\tau\rangle=(\mathbf{1}-\mathbf{G}\mathbf{e}_s)|\phi_k^\tau\rangle \quad (3.14)$$

is the electric field arising from an input beam  $|\phi_k^\tau\rangle$  scattered by the material system. Equation (3.8) gives the electric-field operator in terms of the input photon operators and the noise currents operators. Once the quantum states of the input light beams and of the scattering system are fixed, by using Eq. (3.8) in principle it is possible to compute the electric-field operator in the presence of the scattering system in all the space if the Green tensor  $G_{ij}(\mathbf{r},\mathbf{r}')$  is known. One efficient procedure to calculate Green tensors for complex 2D and 3D scattering objects is described in Ref. [4] and it is based on a

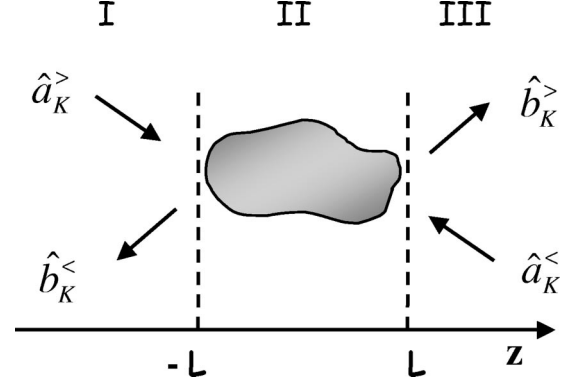


FIG. 1. Scattering geometry and notation.

volume discretization procedure in close analogy with the discrete dipole approximation [31,32]. Recently the method of multipole expansion has been used to calculate 2D Green functions in photonic crystals [33]. A different scheme for the calculation of Green functions for photons propagating in complex dielectric structures based on an extension of the finite-difference time-domain method has been presented by Ward and Pendry [34].

The consistency of the quantization approach described in this section with the equal-time QED commutation relations is proved in Appendix A.

#### IV. QUANTUM-OPTICAL INPUT-OUTPUT RELATIONS

Inside absorbing materials, owing to the presence of noise currents, it is not possible to define space-independent photon operators as in free space [18,29], however, we may attempt to find input and output photon operators outside the scattering system [23]. This would furnish useful input-output quantum-optical relations and it would imply that, just outside the scattering system, the light field, although carrying information on the scattering process, can be quantized as in free space. We proceed by bounding the scattering system with two planes at  $z = \pm L$ , thus separating space in three regions: the left region (I) ( $z < -L$ ), the scattering region (II) ( $-L < z < L$ ), and the right region (III) ( $z > L$ ), as shown in Fig. 1. In the following we will show that it is possible to define space-independent photon operators outside the scattering region. We start from the Dyson equation

$$\mathbf{G}=\mathbf{G}^0-\mathbf{G}^0\mathbf{e}_s\mathbf{G}, \quad (4.1)$$

where  $\mathbf{G}^0$  is the unperturbed free-space Green dyadic, obeying the following equation:

$$(\mathbf{L}+\mathbf{e}_0)\mathbf{G}^0=\mathbf{1}. \quad (4.2)$$

Using the angular plane-wave expansion, the free-space Green tensor can be written as

$$G_{ij}^0(\mathbf{r},\mathbf{r}')=-\frac{i\pi c^2}{\omega}\sum_{\mathbf{K}}\phi_{\mathbf{K},i}^>(\mathbf{r}_>)\phi_{-\mathbf{K},j}^<(\mathbf{r}_<)+\frac{\delta(z-z')}{k^2}\frac{\mathbf{z}\mathbf{z}}{z^2}, \quad (4.3)$$

where  $\mathbf{r}_> = \mathbf{r}(\mathbf{r}_< = \mathbf{r}')$  for  $z > z'$  and  $\mathbf{r}_> = \mathbf{r}'(\mathbf{r}_< = \mathbf{r})$  for  $z < z'$ . Let us analyze the electric-field operator in the regions external to the scattering system (I and III). We start from the contribution (3.10) arising from the solution of the homogeneous wave equation. We introduce the Dyson equation (4.1) into Eq. (3.10),

$$|\hat{\mathbf{E}}_h^+\rangle = |\hat{\mathbf{E}}^{0+}\rangle - \mathbf{G}^0(\mathbf{1} - \mathbf{e}_s \mathbf{G}) \mathbf{e}_s |\hat{\mathbf{E}}^{0+}\rangle. \quad (4.4)$$

Let us now consider the electric-field operator  $\hat{\mathbf{E}}_h^+(\mathbf{r})$  in region III ( $z > L$ ). In this case the free-space Green tensor in Eq. (4.4) appears always with  $z > z'$  and thus can be written simply as

$$G_{ij}^0(\mathbf{r}, \mathbf{r}') = -\frac{i\pi c^2}{\omega} \sum_{\mathbf{K}} \phi_{\mathbf{K},i}^>(\mathbf{r}) \phi_{\mathbf{K},j}^<(\mathbf{r}') \quad (z > z'), \quad (4.5)$$

that in compact notation reads

$$\mathbf{G}^0 = -\frac{i\pi c^2}{\omega} \sum_{\mathbf{K}} |\phi_{\mathbf{K}}^>\rangle \langle \phi_{\mathbf{K}}^>|, \quad (4.6)$$

where we have introduced the following definition:

$$\langle \phi_{\mathbf{K}}^>|\mathbf{r}\rangle \equiv \langle \mathbf{r}|\phi_{-\mathbf{K}}^<|. \quad (4.7)$$

Introducing Eq. (4.6) into Eq. (4.4) and using Eq. (3.14) we obtain

$$|\hat{\mathbf{E}}_h^+\rangle = |\hat{\mathbf{E}}^{0+}\rangle + \frac{i\pi c^2}{\omega} \sum_{\mathbf{K}} |\phi_{\mathbf{K}}^>\rangle \langle \psi_{\mathbf{K}}^>|\mathbf{e}_s|\hat{\mathbf{E}}^{0+}\rangle. \quad (4.8)$$

Following the same steps for  $\hat{\mathbf{E}}_p^+$  we obtain

$$|\hat{\mathbf{E}}_p^+\rangle = i\omega\mu_0 \frac{i\pi c^2}{\omega} \sum_{\mathbf{K}} |\phi_{\mathbf{K}}^>\rangle \langle \psi_{\mathbf{K}}^>|\hat{\mathbf{j}}\rangle. \quad (4.9)$$

By introducing Eq. (3.1) into Eq. (4.8), the total electric-field operator in the region III can be written as

$$\hat{\mathbf{E}}^+(\mathbf{r}, \omega) = i\sqrt{\frac{\hbar\omega}{2\varepsilon_0}} \sum_{\mathbf{K}} [\phi_{\mathbf{K}}^>(\mathbf{r}, \omega) \hat{b}_{\mathbf{K}}^>(\omega) + \phi_{\mathbf{K}}^<(\mathbf{r}, \omega) \hat{a}_{\mathbf{K}}^<(\omega)], \quad (4.10)$$

with the space-independent output photon operators given by

$$\hat{b}_{\mathbf{K}}^> = \hat{b}_{\mathbf{K}}^{h>} + \hat{b}_{\mathbf{K}}^{p>}, \quad (4.11)$$

with

$$\hat{b}_{\mathbf{K}}^{h>} = \hat{a}_{\mathbf{K}}^> + \frac{\pi c^2}{\omega} \sqrt{\frac{2\varepsilon_0}{\hbar\omega}} \langle \psi_{\mathbf{K}}^>|\mathbf{e}_s|\hat{\mathbf{E}}^{0+}\rangle \quad (4.12)$$

and

$$\hat{b}_{\mathbf{K}}^{p>} = \frac{i\pi}{\varepsilon_0} \sqrt{\frac{2\varepsilon_0}{\hbar\omega}} \langle \psi_{\mathbf{K}}^>|\hat{\mathbf{j}}^+\rangle. \quad (4.13)$$

Equation (4.11) with Eqs. (4.12) and (4.13) gives the output photon operator associated with a plane wave of given energy and propagating along a fixed direction (determined by  $\mathbf{K}$  and  $\omega$ ) (see Fig. 1) in terms of the input photon operators and of the noise currents inside the material system. The integrals in Eqs. (4.12) and (4.13) can be explicitly written in the  $\mathbf{r}$  representation, the one in Eq. (4.13) reads

$$\langle \psi_{\mathbf{K}}^>|\hat{\mathbf{j}}^+\rangle = \int \psi_{\mathbf{K}}^<(\mathbf{r}) \cdot \hat{\mathbf{j}}(\mathbf{r}) d\mathbf{r}. \quad (4.14)$$

Analogous results can be obtained for the electric-field operator in region I, that can be written as

$$\hat{\mathbf{E}}^+(\mathbf{r}, \omega) = i\sqrt{\frac{\hbar\omega}{2\varepsilon_0}} \sum_{\mathbf{K}} [\phi_{\mathbf{K}}^>(\mathbf{r}, \omega) \hat{a}_{\mathbf{K}}^>(\omega) + \phi_{\mathbf{K}}^<(\mathbf{r}, \omega) \hat{b}_{\mathbf{K}}^<(\omega)], \quad (4.15)$$

with  $\hat{b}_{\mathbf{K}}^<(\omega) = \hat{b}_{\mathbf{K}}^{h<} + \hat{b}_{\mathbf{K}}^{p<}$  given by

$$\hat{b}_{\mathbf{K}}^{h<} = \hat{a}_{\mathbf{K}}^< + \frac{\pi c^2}{\omega} \sqrt{\frac{2\varepsilon_0}{\hbar\omega}} \langle \psi_{\mathbf{K}}^<|\mathbf{e}_s|\hat{\mathbf{E}}^{0+}\rangle \quad (4.16)$$

and

$$\hat{b}_{\mathbf{K}}^{p<} = \frac{i\pi}{\varepsilon_0} \sqrt{\frac{2\varepsilon_0}{\hbar\omega}} \langle \psi_{\mathbf{K}}^<|\hat{\mathbf{j}}\rangle. \quad (4.17)$$

Equations (4.12) and (4.16) can be further simplified evaluating the integrals. This can be done by using the Lippman-Schwinger equation

$$|\psi\rangle = (\mathbf{1} - \mathbf{G}^0 \mathbf{e}_s) |\psi\rangle. \quad (4.18)$$

By using the angular spectrum representation of the field  $|\psi\rangle$ , defined according to

$$\psi(\mathbf{r}) = \sum_{\mathbf{K}} \psi^{\mathbf{K}}(z) e^{i\mathbf{K}\cdot\mathbf{R}}, \quad (4.19)$$

we can project the Lippman-Schwinger equation as

$$|\psi^{\mathbf{K}}\rangle = |\phi^{\mathbf{K}}\rangle + \frac{i\pi c^2}{\omega} |\phi_{\mathbf{K}}^>(<), \mathbf{K}\rangle \langle \phi_{\mathbf{K}}^>(<)|\mathbf{e}_s|\psi\rangle, \quad (4.20)$$

with  $\tau(\tau')$  depending on which region (I or III) we are considering. Introducing Eq. (3.1) into Eq. (4.12), using Eq. (4.20), and observing that  $\phi_{\mathbf{K},\sigma}^<(\mathbf{K})(L) = \hat{\mathbf{e}}_{\mathbf{K},\sigma}^< \exp[-ik_z L]$ , we obtain

$$\begin{aligned} \langle \psi_{\mathbf{K}}^>|\mathbf{e}_s|\hat{\mathbf{E}}^{0+}\rangle &= \langle \hat{\mathbf{E}}^{0+}|\mathbf{e}_s|\psi_{-\mathbf{K}}^<\rangle \\ &= \sqrt{\frac{\hbar\omega}{2\varepsilon_0}} \sum_{\mathbf{K}'} \alpha_{\mathbf{K}', 2k'_z} A \exp[-ik'_z L] \\ &\quad \times [\{\psi_{\mathbf{K},\sigma}^<(\bar{\mathbf{K}}'(-L)) - \phi_{\mathbf{K}',\sigma}^<(\bar{\mathbf{K}}'(-L))\} e_{\mathbf{K}'}^> \hat{a}_{\mathbf{K}'}^>] \end{aligned}$$

$$+\{\psi_{\bar{\mathbf{K}},\sigma}^{<,\bar{\mathbf{K}}'}(L)-\phi_{\bar{\mathbf{K}},\sigma}^{<,\bar{\mathbf{K}}'}(L)\}e_{K'}^{<,\hat{a}_{K'}^{<}}. \quad (4.21)$$

By using this equation, Eq. (4.12) can be written as

$$\hat{b}_K^{h>} = \sum_{K'} [T_K^{K'} \hat{a}_{K'}^{>} + R_K^{K'} \hat{a}_{K'}^{<}], \quad (4.22)$$

where

$$T_K^{K'} = \frac{e^{-ik'_z L}}{\alpha_{K'}} \psi_{\bar{\mathbf{K}}}^{<,\bar{\mathbf{K}}'}(-L) \cdot \mathbf{e}_{K'}^{>}, \quad (4.23)$$

$$R_K^{K'} = \frac{e^{-ik'_z L}}{\alpha_{K'}} [\psi_{\bar{\mathbf{K}}}^{<,\bar{\mathbf{K}}'}(L) - \phi_{\bar{\mathbf{K}}}^{<,\bar{\mathbf{K}}'}(L)] \cdot \mathbf{e}_{K'}^{<}, \quad (4.24)$$

where  $\bar{\mathbf{K}} \equiv -\mathbf{K}$  and  $\bar{K} = (-\mathbf{K}, \sigma)$ . Analogously, we can obtain for the operators describing output in region I,

$$\hat{b}_K^{h<} = \sum_{K'} [\mathcal{R}_K^{K'} \hat{a}_{K'}^{>} + \mathcal{T}_K^{K'} \hat{a}_{K'}^{<}], \quad (4.25)$$

where

$$\mathcal{R}_K^{K'} = \frac{e^{-ik'_z L}}{\alpha_{K'}} [\psi_{\bar{\mathbf{K}}}^{>,\bar{\mathbf{K}}'}(-L) - \phi_{\bar{\mathbf{K}}}^{>,\bar{\mathbf{K}}'}(-L)] \cdot \mathbf{e}_{K'}^{>}, \quad (4.26)$$

$$\mathcal{T}_K^{K'} = \frac{e^{-ik'_z L}}{\alpha_{K'}} \psi_{\bar{\mathbf{K}}}^{>,\bar{\mathbf{K}}'}(L) \cdot \mathbf{e}_{K'}^{<}. \quad (4.27)$$

The obtained quantum-optical input-output relations relate the output operators  $\hat{\mathbf{b}}_K \equiv (\hat{b}_K^{>}, \hat{b}_K^{<})$  to the input photon operators  $\hat{\mathbf{a}}_K \equiv (\hat{a}_K^{>}, \hat{a}_K^{<})$  and to the noise currents  $\hat{\mathbf{j}}(\mathbf{r})$  of the scattering system, according to

$$\hat{\mathbf{b}}_K = \sum_{K'} \mathbf{S}_K^{K'} \hat{\mathbf{a}}_{K'} + \hat{\mathbf{F}}_K, \quad (4.28)$$

where  $\mathbf{S}_K^{K'}$  is a  $2 \times 2$  scattering matrix ( $S$  matrix),

$$\mathbf{S}_K^{K'} = \begin{pmatrix} T_K^{K'} & R_K^{K'} \\ \mathcal{R}_K^{K'} & \mathcal{T}_K^{K'} \end{pmatrix}, \quad (4.29)$$

and  $\hat{\mathbf{F}}_K$  is a two-dimensional quantum noise vector,

$$\hat{\mathbf{F}}_K = \frac{i\pi}{\varepsilon_0} \sqrt{\frac{2\varepsilon_0}{\hbar\omega}} (\langle \psi_K^{>} | \hat{\mathbf{j}} \rangle, \langle \psi_K^{<} | \hat{\mathbf{j}} \rangle).$$

If the quantum state of input radiation and of the material system is known, any output photon correlation can be directly calculated by using these relations provided the classical light modes  $\psi_{\bar{\mathbf{K}}}^{<}$  have been computed. Light modes for specific complex structures can be calculated using Eq. (3.14) according to the scheme described in Ref. [5]. We

observe that, since we have not assumed any translation symmetry for our scattering system, in principle all the light modes  $\psi_{\bar{\mathbf{K}}}^{<}$  arising from all possible input fields  $K'$  are expected to contribute to output waves propagating along a given direction determined by  $\mathbf{K}$  and  $\omega$ . Instead, due to reciprocity, Eqs. (4.12) and (4.13) have a simpler structure showing that  $\hat{b}_K^{>}$  depends on  $\hat{\mathbf{E}}^{0+}$  and  $\hat{\mathbf{j}}^+$  only via the reciprocal mode  $\psi_{\bar{\mathbf{K}}}^{<}$ . The obtained input-output relations (4.28) are based on the angular-spectrum representation. As it is well known, this representation describes explicitly also the evanescent waves [6] that appears for  $K > k$ . These relations (4.28) hold also for evanescent fields and define naturally output photon operators associated with evanescent waves. With the improvement in techniques based on measurement and control of evanescent waves, these relations should find application for the analysis of evanescent nonclassical fields, e.g., arising from the scattering of nonclassical input fields by nanometric objects.

## V. COMMUTATION RELATIONS

The expansion in input and output photon operators performed above is consistent only if the output operators are true photon operators obeying Bosonic commutation rules. Let us start looking at the commutator for the particular term of the rightward output operator. By using Eqs. (2.3) and (4.13), we obtain

$$[\hat{b}_K^{p>}(\omega), \hat{b}_{K'}^{p>\dagger}(\omega')] = \frac{\pi}{\varepsilon_0} A_{K,K'}(\omega) \delta(\omega - \omega'), \quad (5.1)$$

with

$$A_{K,K'} = \int d\mathbf{r} [\psi_{\bar{\mathbf{K}}}^{<}(\mathbf{r}) \cdot \mathbf{J}_{\bar{\mathbf{K}}'}^{<*}(\mathbf{r}) + \mathbf{J}_{\bar{\mathbf{K}}}^{<}(\mathbf{r}) \cdot \psi_{\bar{\mathbf{K}}'}^{<*}(\mathbf{r})], \quad (5.2)$$

where  $\mathbf{J}(\omega) = -i\omega\varepsilon_0\chi\psi(\omega)$ . We observe that  $A_{K,K}$  is the power loss of mode  $\psi_{\bar{\mathbf{K}}}^{<}(\omega)$  due to the scattering system. From the Maxwell equations, following the same steps as for the derivation of the Poynting theorem, we find that

$$A_{K,K'} + \Phi_{K,K'} = 0, \quad (5.3)$$

with

$$\Phi_{K,K'} = \oint [\psi_{\bar{\mathbf{K}}}^{<}(\mathbf{r}) \times \mathbf{H}_{\bar{\mathbf{K}}'}^{<*}(\mathbf{r}) + \psi_{\bar{\mathbf{K}}'}^{<*}(\mathbf{r}) \times \mathbf{H}_{\bar{\mathbf{K}}}^{<}(\mathbf{r})] \cdot \mathbf{n} da, \quad (5.4)$$

where  $\mathbf{H} = (1/i\omega\mu_0)\nabla \times \psi$  is the corresponding magnetic field, the integration is over a surface bounding the scattering system and  $\mathbf{n}$  is the unit vector normal to the surface.  $-\Phi_{K,K}$  is the real power flowing into the scattering system. Equation (5.3) is a compact form for the Poynting theorem ( $K=K'$ ) and for the Lorentz reciprocity theorem ( $K \neq K'$ ). By manipulating the vector products, Eq. (5.4) can be rewritten as

$$\Phi_{K,K'} = \frac{i}{\omega\mu_0} \int \left[ \boldsymbol{\psi}_K^<(\mathbf{r}) \cdot \frac{\partial}{\partial \mathbf{n}} \boldsymbol{\psi}_{K'}^{<*}(\mathbf{r}) - \boldsymbol{\psi}_{K'}^{<*}(\mathbf{r}) \cdot \frac{\partial}{\partial \mathbf{n}} \boldsymbol{\psi}_K^<(\mathbf{r}) \right] da. \quad (5.5)$$

This surface integral can be evaluated by choosing as bounding surface the two planes at  $z = \pm L$  and by using the angular-spectrum representation of the fields (4.19). The gradients in the direction normal to the surfaces can be easily evaluated by using Eq. (4.20). We obtain

$$\Phi_{K,K'} = \Phi_{K,K'}^{\text{out}} - \Phi_{K,K'}^{\text{in}}, \quad (5.6)$$

with

$$\Phi_{K,K'}^{\text{out}} = \frac{\varepsilon_0}{\pi} \sum_Q [R_K^Q R_{K'}^{Q*} + T_K^Q T_{K'}^{Q*}], \quad (5.7)$$

$$\Phi_{K,K'}^{\text{in}} = \frac{\varepsilon_0}{\pi} \delta_{K,K'}. \quad (5.8)$$

Using Eq. (4.22), we also directly obtain

$$[\hat{b}_K^{h>}(\omega), \hat{b}_{K'}^{h>\dagger}(\omega')] = \frac{\pi}{\varepsilon_0} \Phi_{K,K'}^{\text{out}}(\omega) \delta(\omega - \omega'). \quad (5.9)$$

Summing Eq. (5.1) and Eq. (5.9), the Boson commutation rules for the output operators are thus readily obtained. Result of this is that the *commutation relations for the output operators are determined by energy conservation ( $K=K'$ ) and by reciprocity ( $K \neq K'$ )*. In particular, reciprocity ensures the independence of output operators with different wave vector or polarization ( $[\hat{b}_K^{h>}, \hat{b}_{K'}^{h>\dagger}] = 0$  for  $K \neq K'$ ). It would be violated if output operators with different wave vector or polarization are not independent as much as the input operators are. So far we have discussed only the commutation relations for the output photon operators. The equal-time QED commutation relations between the fundamental fields are shown in Appendix A.

## VI. LIGHT EMISSION AND ELECTRIC-FIELD FLUCTUATIONS

In this section we analyze the fluctuation properties of the electromagnetic field in presence of absorbing and/or emitting media and present some examples of light propagation in nonequilibrium.

Let us start considering vacuum fluctuations in presence of a scattering system. As it is well known, vacuum fluctuations play a fundamental role in quantum-optical processes [37]. By using Eq. (3.8) and the relation (A4) we obtain

$$\langle \hat{\mathbf{E}}_i(\mathbf{r}_1, \omega) \hat{\mathbf{E}}_j(\mathbf{r}_2, \omega') \rangle_{0,0} = S_{ij}^0(\mathbf{r}_1, \mathbf{r}_2, \omega) \delta(\omega - \omega'), \quad (6.1)$$

with

$$S_{ij}^0(\mathbf{r}_1, \mathbf{r}_2, \omega) = -(\hbar \omega^2 / \varepsilon_0 \pi c^2) G_{ij}^I(\mathbf{r}_1, \mathbf{r}_2, \omega). \quad (6.2)$$

In Eq. (6.1),  $\langle \rangle_{a,b}$  indicates the expectation value, where  $(a,b)$  labels respectively the state of input light and the state of the material system. In this case  $(0,0)$  indicates the vacuum state for both the Hilbert spaces. Equation (6.2) agrees with results obtained by applying the fluctuation-dissipation theorem [15].

Let us now consider a scattering system with an effective uniform temperature  $T$  embedded in a vacuum at *zero* temperature. By using Eq. (3.8) we obtain

$$\langle \hat{\mathbf{E}}_i^-(\mathbf{r}_1, \omega) \hat{\mathbf{E}}_j^+(\mathbf{r}_2, \omega') \rangle_{0,T} = W_{ij}(\mathbf{r}_1, \mathbf{r}_2, \omega) \delta(\omega - \omega'), \quad (6.3)$$

with

$$W_{ij}(\mathbf{r}_1, \mathbf{r}_2, \omega) = N(\omega, T) (\hbar \omega^2 / \varepsilon_0 \pi c^2) \tilde{A}(\mathbf{r}_1, \mathbf{r}_2, \omega). \quad (6.4)$$

where  $\tilde{A}(\mathbf{r}_1, \mathbf{r}_2, \omega)$  is defined in Appendix A. Equation (6.4) is very similar to the expression used in Ref. [6] to calculate the cross-spectral density tensor of the near field thermally emitted into free space by an opaque planar source. Using Eq. (A4), Eq. (6.4) can be written in the form

$$\mathbf{W}(\mathbf{r}, \mathbf{r}', \omega) = \bar{N}(\omega, T) \left[ \mathbf{S}^0(\mathbf{r}, \mathbf{r}', \omega) - \frac{\hbar \omega}{2\varepsilon_0} \tilde{\boldsymbol{\rho}}(\mathbf{r}, \mathbf{r}', \omega) \right]. \quad (6.5)$$

This equation establishes a general relationship between the spatial variations of the second-order coherence tensors for vacuum fluctuations and spontaneous light emission. We observe that, while vacuum fluctuations originate from both the scattering system and the input light modes, light emission in a zero-temperature free space comes only from the scattering system. This explains why the spatial variation of the tensor describing light emission can be obtained by subtracting from the contribution due to the vacuum fluctuation the contribution originating from the input light modes  $\tilde{\boldsymbol{\rho}}(\mathbf{r}, \mathbf{r}', \omega)$  and eventually reflected by the thermal source.

The noise properties of the electromagnetic field are manifested by electric-field fluctuation spectrum in the absence of any input signal. Let us consider a material system at a given uniform temperature. The electric-field correlation spectrum is defined by

$$\begin{aligned} \langle \hat{\mathbf{E}}(\mathbf{r}, \omega) \hat{\mathbf{E}}(\mathbf{r}', \omega') \rangle_{0,T} &= \langle \hat{\mathbf{E}}_h(\mathbf{r}, \omega) \hat{\mathbf{E}}_h(\mathbf{r}', \omega') \rangle_0 \\ &\quad + \langle \hat{\mathbf{E}}_p(\mathbf{r}, \omega) \hat{\mathbf{E}}_p(\mathbf{r}', \omega') \rangle_T \\ &= \mathbf{S}(\mathbf{r}, \mathbf{r}', \omega) \delta(\omega - \omega'). \end{aligned} \quad (6.6)$$

By inserting the expression for the electric-field operator derived in Sec. III, we obtain

$$\begin{aligned} \langle \hat{\mathbf{E}}_h(\mathbf{r}, \omega) \hat{\mathbf{E}}_h(\mathbf{r}', \omega') \rangle_0 &= \langle \hat{\mathbf{E}}_h^+(\mathbf{r}, \omega) \hat{\mathbf{E}}_h^-(\mathbf{r}', \omega') \rangle_0 \\ &= \frac{\hbar \omega}{2\varepsilon_0} \tilde{\boldsymbol{\rho}}(\mathbf{r}, \mathbf{r}', \omega) \delta(\omega - \omega'), \end{aligned} \quad (6.7)$$

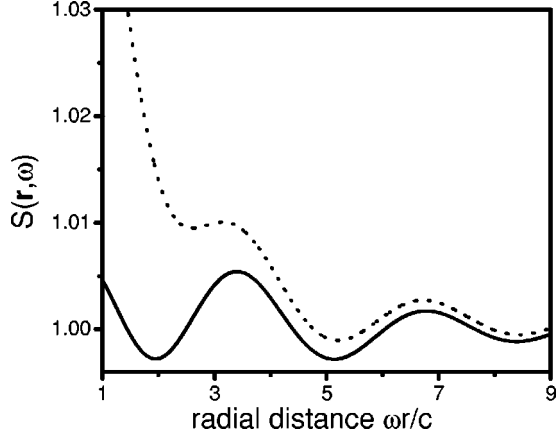


FIG. 2. Normalized (with respect to free space) electric-field fluctuations for a resonant-point scatterer at zero temperature (continuous line) and at a given effective temperature (dotted line) as a function of distance from the scatterer. Parameters are given in the text.

$$\langle \hat{E}_p(\mathbf{r}, \omega) \hat{E}_p(\mathbf{r}', \omega') \rangle_T = \frac{2N(\omega, T) + 1}{N(\omega, T)} \times \mathbf{W}(\mathbf{r}, \mathbf{r}', \omega) \delta(\omega - \omega'), \quad (6.8)$$

hence Eq. (6.6) can be written as

$$\mathbf{S}(\mathbf{r}, \mathbf{r}', \omega) = \mathbf{S}^0(\mathbf{r}, \mathbf{r}', \omega) + 2\mathbf{W}(\mathbf{r}, \mathbf{r}', \omega). \quad (6.9)$$

We observe that both  $\mathbf{S}^0$  and  $\mathbf{W}$  for a specific system can be directly calculated once the Green tensor has been derived. The power spectrum  $S(\mathbf{r}, \omega)$  of the electric-field fluctuations at position  $\mathbf{r}$  is obtained by taking the trace of Eq. (6.6). These power spectra are usually obtained using the fluctuation-dissipation theorem [18,19]. Fluctuation-dissipation theorems have also been derived for amplifying media [38,39]. However, this approach cannot be used when the whole system is not in thermal equilibrium as in the present example. In this case we are considering an attenuating or amplifying medium at a given effective temperature embedded in free space at zero temperature. In Fig. 2, we display the electric-field fluctuations  $S(\mathbf{r}, \omega)$  as a function of the radial distance for a pointlike scattering object embedded in free space. In Appendix B, the Green tensor for this elementary scattering system is derived. Figure 2 shows  $S(\mathbf{r}, \omega)$  (normalized with respect to the free-space value) for  $T=0$  and for an effective temperature such that  $N(\omega, T)=3$ . We have considered a point scatterer of radius  $a=15$  nm with complex permittivity  $\varepsilon=6+0.8i$ . The wavelength of the radiation is 600 nm.

Let us now analyze another nonequilibrium physical situation. We consider a scattering system with an effective uniform temperature  $T_1$  with mode occupation  $N_1$  embedded in a *thermal* free space that is the cavity of a black body at temperature  $T_2$  and mode occupation  $N_2$  with walls very far from the scattering system. In this case the electric-field cross-spectral density tensor  $\mathbf{W}'$ ,

$$\langle \hat{\mathbf{E}}_i^-(\mathbf{r}_1, \omega) \hat{\mathbf{E}}_j^+(\mathbf{r}_2, \omega') \rangle_{T_2, T_1} = W'_{ij}(\mathbf{r}_1, \mathbf{r}_2, \omega) \delta(\omega - \omega'), \quad (6.10)$$

is given by the following expression:

$$W'_{ij}(\mathbf{r}_1, \mathbf{r}_2, \omega) = N_1 S_{ij}^0(\mathbf{r}_1, \mathbf{r}_2, \omega) + \frac{\hbar \omega}{2\varepsilon_0} \rho_{ij}(\mathbf{r}_1, \mathbf{r}_2, \omega) [N_2 - N_1]. \quad (6.11)$$

We observe that the spatial variations of this correlation function change continuously as a function of  $T_1$  and  $T_2$ . At equilibrium ( $T_1=T_2$ ) the spatial behavior of  $\mathbf{W}'$  coincides with that of  $\mathbf{G}^I$  and hence with that of the tensor  $\mathbf{S}$  describing vacuum fluctuations. As expected  $\mathbf{G}^I$  describes the electromagnetic-field fluctuations at equilibrium [15]. The light intensity as a function of frequency is proportional to  $I'(\mathbf{r}, \omega) = \text{Tr} \mathbf{W}'(\mathbf{r}, \mathbf{r}, \omega)$ . We obtain

$$I'(\mathbf{r}, \omega) = N_1 \text{Tr} \mathbf{S}^0(\mathbf{r}, \mathbf{r}, \omega) + \frac{\hbar \omega}{2\varepsilon_0} \rho(\mathbf{r}, \omega) [N_2 - N_1], \quad (6.12)$$

where  $\rho(\mathbf{r}, \omega) = \text{Tr} \tilde{\rho}(\mathbf{r}, \mathbf{r}, \omega)$ ; as it can be inferred from Eq. (A3),  $\rho(\mathbf{r}, \omega)$  describes the local optical density of states (DOS). It gives the intensity of light at  $\mathbf{r}$  due to incoherent illumination, i.e., with input light modes arriving from all the spatial directions and it is currently used to characterize the optical properties of PBG structures [4] and more generally of dielectric systems [35,36]. Before presenting some numerical results, we observe that when  $N_1$  equals  $N_2$ , a situation of thermal equilibrium is recovered and the spatial variation of light intensity is the same of vacuum fluctuations and is determined by the trace of the imaginary part of the Green tensor as prescribed by the fluctuation-dissipation theorem. Out of equilibrium the fluctuation-dissipation theorem does not hold. If  $N_1=0$ , which means that the medium is in its ground state and does not emit light, the spatial variation of  $I'(\mathbf{r}, \omega)$  is determined by the local optical density of states  $\rho(\mathbf{r}, \omega)$ . In the opposite limit  $N_2=0$ , there is no input light and the spatial variation of  $I'(\mathbf{r}, \omega)$  describes the emission pattern of the medium that is given by the trace of Eq. (6.4).

Figure 3 displays  $I'(\mathbf{r}, \omega) / [(N_1 + N_2) \text{Tr} \mathbf{G}_0^I]$  for the same pointlike scattering object of Fig. 2. We consider different ratios  $N_2/N_1$ . Figure 3(a) obtained with  $N_2/N_1=0$  displays the emission pattern of the pointlike scatterer. Figure 3(c) calculated at equilibrium ( $N_1=N_2$ ) displays  $\text{Tr} \mathbf{G}^I / \text{Tr} \mathbf{G}_0^I$ . Fig. 3(e) displays the normalized local DOS. The other two panels describe intermediate situations. We point out that the oscillations observed in Figs. 3(b)–3(e) originate from the interference between the input and the reflected light fields. These oscillations are absent in Fig. 3(a) because in this case there is only emission from the scattering object. Figure 3 shows that oscillations increase when increasing the ratio  $N_2/N_1$ . This is the consequence of the definite phase relation between the input and the scattered lights (the input and the scattered lights are proportional to  $N_2$ ), on the contrary the emitted light  $\propto N_1$  does not interfere with input light. We also

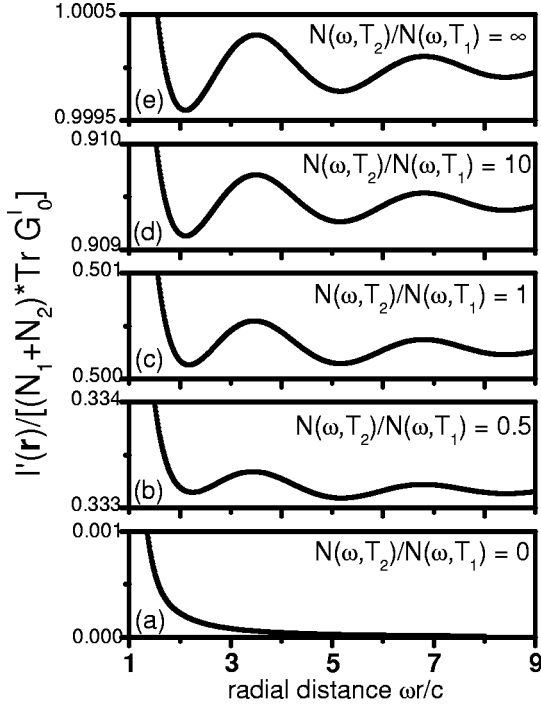


FIG. 3. Normalized light intensity  $I'(\mathbf{r}, \omega) / [(N_1 + N_2) \text{Tr} \mathbf{G}_0^I]$  as a function of distance from a resonant-point scatterer under different mode occupations  $N_1$  and  $N_2$ . Parameters are given in the text.

observe that Figs. 3(c) and 3(e) display a different spatial behavior showing that in the presence of absorption the well-known relationship between the Green tensor and the local DOS,

$$\rho(\mathbf{r}, \omega) = -\frac{2\omega}{\pi c^2} \text{Tr} \mathbf{G}^I(\mathbf{r}, \mathbf{r}, \omega) \quad (6.13)$$

is not correct.

## VII. CONCLUSIONS

In conclusion, we have presented a general quantum theory of light scattering for 3D systems of arbitrary geometry, providing a unified basis for analyzing a large class of optical processes where quantum and/or thermal fluctuations play a role. We have derived general 3D quantum-optical input-output relations providing the output photon operators in terms of the input photon operators and of the noise currents of the scattering system. These relations hold also for photon operators associated with evanescent fields and thus can be applied to the analysis of evanescent nonclassical fields, e.g., arising from the scattering of nonclassical input light by nanometric objects. The theory puts forward the connection between general theorems of classical electrodynamics and commutation relations for the output photon operators carrying all the information on the scattering and/or emission process. We have shown that this theory satisfies QED commutation rules by using a novel relationship between vacuum and thermal fluctuations. Applications involving scattering from complex nanometric scattering objects

are under current development.

## ACKNOWLEDGMENT

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## APPENDIX A: QED COMMUTATION RELATIONS

In order to ensure overall consistency of this treatment, we show how fundamental equal-time commutation relations of QED are preserved. This consistency check has been proved for a quite general three-dimensional dielectric with a local and scalar permittivity, not including the homogeneous solution of the electric-field operator [25], i.e., by expressing the electric-field operator via the Green tensor as a function of only the noise currents. Here we generalize this result showing that equal-time commutation relations of QED are preserved also for more general systems that can be anisotropic or even driven by a nonlocal susceptibility. Moreover, we also include the homogeneous solution of Eq. (2.1), thus considering explicitly the scattered fields from bounded scattering systems. Let us consider the (equal-time) commutation relations between the fundamental fields  $\hat{\mathbf{E}}(\mathbf{r}, t)$  and  $\hat{\mathbf{B}}(\mathbf{r}, t)$ . Using the expression for the homogeneous electric-field operator and that  $i\omega\hat{\mathbf{B}}^+(\omega) = \nabla \times \hat{\mathbf{E}}^+(\omega)$ , we obtain

$$[\hat{E}_i(\mathbf{r}), \hat{B}_l(\mathbf{r}')] = \frac{i\hbar}{2\epsilon_0} \epsilon_{lmj} \partial_m^{r'} \int_0^\infty d\omega \left[ \tilde{\rho}_{i,j}(\mathbf{r}, \mathbf{r}') + \frac{2\omega}{\pi c^2} \tilde{\mathbf{A}}_{ij}(\mathbf{r}, \mathbf{r}') \right] - \text{c.c.}, \quad (A1)$$

where

$$\tilde{\mathbf{A}} = \mathbf{G}e^I \mathbf{G}^* \quad (A2)$$

and

$$\tilde{\rho}_{i,j}(\mathbf{r}, \mathbf{r}') = \sum_{\tau, K} \psi_{K,i}^\tau(\mathbf{r}) \psi_{K,j}^{\tau*}(\mathbf{r}'). \quad (A3)$$

Equation (A1) can be simplified using the following relation:

$$\frac{\pi c^2}{2\omega} \tilde{\rho}_{i,j}(\mathbf{r}, \mathbf{r}') = -\mathbf{G}_{ij}^I(\mathbf{r}, \mathbf{r}') - \tilde{\mathbf{A}}_{ij}(\mathbf{r}, \mathbf{r}'). \quad (A4)$$

This equation can be proved using the mode expansion (4.3) of  $\mathbf{G}^0$  and applying the Dyson equation. Equation (A4) has been demonstrated for particular cases [21,23]. Its general derivation will be presented elsewhere. By using Eq. (A4), Eq. (A1) reduces to

$$[\hat{E}_i(\mathbf{r}), \hat{B}_l(\mathbf{r}')] = -\frac{i\hbar}{\pi\epsilon_0} \epsilon_{lmj} \partial_m^{r'} \int_0^\infty d\omega \frac{\omega}{c^2} G_{ij}^I(\mathbf{r}, \mathbf{r}', \omega) - \text{c.c.} \quad (A5)$$



Furthermore, from Eq. (3.7) and the relation  $\chi^*(\omega) = \chi(-\omega)$  it follows that  $G_{ij}^*(\mathbf{r}, \mathbf{r}', \omega) = G_{ij}(\mathbf{r}, \mathbf{r}', -\omega)$ , Eq. (A5) becomes

$$[\hat{E}_i(\mathbf{r}), \hat{B}_l(\mathbf{r}')] = -\frac{\hbar}{\pi \epsilon_0} \epsilon_{lmj} \partial_m^{r'} \int_{-\infty}^{\infty} d\omega \frac{\omega}{c^2} G_{ij}(\mathbf{r}, \mathbf{r}', \omega). \quad (\text{A6})$$

Although, owing to generalizations, the starting point (A1) was quite different from Eq. (26) of Ref. [24], after using Eq. (A4) and some manipulation we arrived at Eq. (A6) that coincides with the corresponding findings in Refs. [11,24]. From now the canonical commutation relations can be demonstrated using a machinery analogous to that of Ref. [11]. In particular equal-time commutation relations of QED are preserved if the following relation holds:

$$\epsilon_{lmj} \partial_m^{r'} \int_{-\infty}^{\infty} d\omega \frac{\omega}{c^2} G_{ij}(\mathbf{r}, \mathbf{r}', \omega) = -i\pi \epsilon_{lmj} \partial_m^{r'} \delta_{ij} \delta(\mathbf{r} - \mathbf{r}'). \quad (\text{A7})$$

For the sake of completeness and also because we adopt a medium susceptibility with a more complex structure, in the following we provide a concise demonstration of Eq. (A7).

First we observe that the Kramers-Kronig relations imply that the causal-complex-valued susceptibility tensor  $\chi_{ij}(\mathbf{r}, \mathbf{r}', \omega)$  is a holomorphic function of  $\omega$  in the upper complex plane. Moreover, Kramers-Kronig relations imply that for  $|\omega| \rightarrow \infty$ ,  $\chi_{ij}(\mathbf{r}, \mathbf{r}', \omega) \rightarrow 0$  at least as  $\omega^{-1}$ . Also the Green tensor for causality requirements is a holomorphic function of  $\omega$  in the upper complex plane. This can also be derived explicitly from applying iteratively the Dyson equation and observing that both  $\chi_{ij}$  and  $\mathbf{G}^0$  are holomorphic in the upper complex plane. The analytical properties of  $\mathbf{G}^0$  can be directly inspected, with its analytical expression being known. The free-space Green tensor can be expanded in plane waves as

$$G_{ij}^0(\mathbf{r}, \mathbf{r}', \omega) = \frac{1}{(2\pi)^3} \int G_{ij}^0(\mathbf{p}, \omega) e^{i\mathbf{p} \cdot (\mathbf{r} - \mathbf{r}')} d\mathbf{p}, \quad (\text{A8})$$

with

$$G_{ij}^0(\mathbf{p}, \omega) = \frac{1}{k^2 - p^2} \left( \delta_{ij} - \frac{p_i p_j}{p^2} \right) + \frac{1}{k^2} \frac{p_i p_j}{p^2}. \quad (\text{A9})$$

From these expansion it follows that for  $|\omega| \rightarrow \infty$ ,  $G_{ij}^0(\mathbf{r}, \mathbf{r}', \omega)$  approaches zero as  $\omega^{-2}$ . We note that  $G_{ij}^0(\mathbf{r}, \mathbf{r}', \omega)$  [see Eq. (A9)] is singular at  $\omega=0$ .

By introducing the Dyson equation into the integral on the right-hand side (rhs) of Eq. (A11), the expression inside the integral becomes

$$\frac{\omega}{c^2} \mathbf{G} = \frac{\omega}{c^2} \mathbf{G}^0 + \frac{\omega}{c^2} \sum_{n=1}^{\infty} [\mathbf{G}^0 \mathbf{e}_s]^n \mathbf{G}^0, \quad (\text{A10})$$

where we have developed by iteration the Dyson equation. Owing to the last term in Eq. (A9), the two terms on the rhs

of Eq. (A10) are singular at  $\omega=0$ . Nevertheless, these singularities, that have to be treated as principal values, does not contribute to the integral being odd functions ( $\propto \omega^{-(2n+1)}$ ) of  $\omega$ . Now we observe that the summation on the left-hand side of Eq. (A10) for  $|\omega| \rightarrow \infty$  approaches zero at least as  $\omega^{-2}$ , thus this summation does not contribute to the integral as can be evaluated by performing the integration on the upper complex plane. As a consequence, we obtain

$$\int_{-\infty}^{\infty} d\omega \frac{\omega}{c^2} G_{ij}(\mathbf{r}, \mathbf{r}', \omega) = \int_{-\infty}^{\infty} d\omega \frac{\omega}{c^2} G_{ij}^0(\mathbf{r}, \mathbf{r}', \omega). \quad (\text{A11})$$

The expression for the  $G^0$  in the real space is

$$G_{ij}^0(\mathbf{r}, \mathbf{r}', \omega) = -\left( \delta_{ij} - \frac{1}{k^2} \partial_i^r \partial_j^{r'} \right) g^0(\mathbf{r} - \mathbf{r}', \omega), \quad (\text{A12})$$

where

$$g^0(\mathbf{r} - \mathbf{r}', \omega) = \frac{e^{ik|\mathbf{r} - \mathbf{r}'|}}{4\pi|\mathbf{r} - \mathbf{r}'|}.$$

Inserting Eq (A12) in Eq. (A6) and recalling that  $\epsilon_{lmj} \partial_m^{r'} \partial_j^{r'} \{ \dots \} = 0$  we obtain

$$[\hat{E}_i(\mathbf{r}), \hat{B}_l(\mathbf{r}')] = \frac{\hbar}{\pi \epsilon_0} \epsilon_{lmj} \partial_m^{r'} \delta_{ij} \int_{-\infty}^{\infty} d\omega \frac{\omega}{c^2} g^0(\mathbf{r} - \mathbf{r}', \omega). \quad (\text{A13})$$

Using the known relation [40]

$$\int_{-\infty}^{\infty} d\omega \frac{\omega}{c^2} g^0(\mathbf{r} - \mathbf{r}', \omega) = i\pi \delta(\mathbf{r} - \mathbf{r}'), \quad (\text{A14})$$

we finally obtain

$$[\hat{E}_i(\mathbf{r}), \hat{B}_l(\mathbf{r}')] = -\frac{i\hbar}{\epsilon_0} \epsilon_{lmj} \partial_m^{r'} \delta_{ij} \delta(\mathbf{r} - \mathbf{r}'). \quad (\text{A15})$$

Similarly, it can be shown that

$$[\hat{E}_i(\mathbf{r}), \hat{E}_l(\mathbf{r}')] = 0,$$

$$[\hat{B}_i(\mathbf{r}), \hat{B}_l(\mathbf{r}')] = 0.$$

We also point out that Eq. (A7) proved here is also the condition for obtaining the correct commutation relations for the potentials and canonically conjugated momenta [11,24].

## APPENDIX B: THE GREEN TENSOR FOR A POINTLIKE SCATTERING OBJECT

Let us consider an absorbing pointlike scattering object. It can be regarded as the building block of much more complicated scattering objects. It has been shown how to calculate the Green tensor of complex nanopatterned scattering objects by discretizing them in terms of these building blocks [4]. Following the approach by de Vries *et al.* [35], it is possible to obtain an analytical expression for the Green tensor of this very simple 3D system.

As it is well known, the Green tensor at  $r=0$  has a singular behavior. Performing a regularization procedure [35]

described at the end of this Appendix, we obtain the free-space-regularized Green tensor that we use as starting point for subsequent calculations. From the Dyson equation, after simple algebra, we obtain the following expression for the Green tensor in the presence of a point scatterer,

$$\tilde{\mathbf{G}}(r, \omega) = [1 - t(\omega)\tilde{\mathbf{G}}^0(r=0, \omega)]\tilde{\mathbf{G}}^0(r, \omega). \quad (\text{B1})$$

Now we can derive the tensor defined in Eq. (A2). As shown in Sec. VI, this tensor describes the emission pattern from the medium at uniform temperature. From Eq. (A2) and using Eq. (B1), we obtain

$$\begin{aligned} \text{Tr} \mathbf{A}(\mathbf{r}, \mathbf{r}, \omega) &= \frac{\omega^2}{c^2} \chi^I(\omega) |1 - t(\omega)| \\ &\times |\tilde{\mathbf{G}}^0(r=0, \omega)|^2 \text{Tr}[\tilde{\mathbf{G}}^0(r, \omega)]^2 \Delta. \end{aligned} \quad (\text{B2})$$

Let us now consider the regularization procedure. We start from the free-space Green tensor, Eq. (A12). Calculating the gradients, Eq. (A12) can be written as

$$\mathbf{G}^0(\mathbf{r}, \mathbf{r}', \omega) = -\frac{e^{ikr}}{4\pi r} [P(ikr)\mathbf{1} + Q(ikr)\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}] + \frac{\delta(\mathbf{r})}{3k^2} \mathbf{1}, \quad (\text{B3})$$

where  $r = |\mathbf{r} - \mathbf{r}'|$ ,  $\hat{\mathbf{r}} = \mathbf{r}/r$  and  $\mathbf{1}$  is the identity operator. We have also defined the functions

$$P(z) = \left(1 - \frac{1}{z} + \frac{1}{z^2}\right), \quad Q(z) = \left(-1 + \frac{3}{z} - \frac{3}{z^2}\right). \quad (\text{B4})$$

The Green tensor can be separated into the transverse and longitudinal parts as follows:

$$\mathbf{G}_T^0(\mathbf{r}, \mathbf{r}', \omega) = -\frac{1 - 3\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}}{4\pi k^2 r^3} - \frac{e^{ikr}}{4\pi r} [P(ikr)\hat{\mathbf{1}} + Q(ikr)\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}], \quad (\text{B5})$$

and

$$\mathbf{G}_L^0(\mathbf{r}, \mathbf{r}', \omega) = \frac{1 - 3\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}}{4\pi k^2 r^3} + \frac{\delta(\mathbf{r})}{3k^2} \hat{\mathbf{1}}. \quad (\text{B6})$$

As can be observed, the Green tensor at  $r=0$  has a singular behavior. A regularization procedure is needed. We follow the regularization procedure described by de Vries *et al.* [35]. As it can be observed, the singularities of the transverse and longitudinal parts of the Green tensor differ, so we need two different regularization procedures. In order to moderate the large- $p$  behavior of these function, we multiply their Fourier transform in  $\mathbf{p}$  space respectively by  $\Lambda_T^2/(\Lambda_T^2 + p^2)$  and  $\Lambda_L^4/(\Lambda_L^4 + p^4)$ . To alter the zeroth-order dynamics as little as possible, one has to take the cutoff momenta  $\Lambda_T$  and  $\Lambda_L$  sufficiently large as compared to  $\omega/c$ . The so-obtained regularized Green tensor is given by

$$\begin{aligned} \tilde{\mathbf{G}}_T^0(\mathbf{r}, \mathbf{r}', \omega) &= -\frac{1 - 3\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}}{4\pi k^2 r^3} \left[ -\frac{e^{ikr}}{4\pi r} [P(ikr)\hat{\mathbf{1}} + Q(ikr)\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}] \right. \\ &\quad \left. - \frac{e^{-\Lambda_T r}}{4\pi r} [P(-\Lambda_T r)\hat{\mathbf{1}} + Q(-\Lambda_T r)\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}] \right] \end{aligned} \quad (\text{B7})$$

and

$$\begin{aligned} \tilde{\mathbf{G}}_L^0(\mathbf{r}, \mathbf{r}', \omega) &= \frac{1 - 3\hat{\mathbf{r}} \otimes \hat{\mathbf{r}}}{4\pi k^2 r^3} \{1 - e^{-\Lambda_L r} [\cos \Lambda_L r + \Lambda_L r (\cos \Lambda_L r \\ &\quad + \sin \Lambda_L r)]\} + \frac{\Lambda_L^2 e^{-\Lambda_L r} \sin \Lambda_L r}{2\pi k^2 r} \hat{\mathbf{r}} \otimes \hat{\mathbf{r}}. \end{aligned} \quad (\text{B8})$$

It can be seen that the regularized Green functions converge exponentially to their unregularized counterparts. In fact, we retrieve the original transverse and longitudinal Green-tensors by letting  $\Lambda_T, \Lambda_L \rightarrow \infty$ . After this regularization procedure the free-space Green tensor at  $r=0$  is no more singular and reads

$$\begin{aligned} \tilde{\mathbf{G}}^0(r=0, \omega) &= \tilde{\mathbf{G}}_T^0(r=0, \omega) + \tilde{\mathbf{G}}_L^0(r=0, \omega) \\ &= \left( \frac{\Lambda_L^3}{6\pi k^2} - \frac{\Lambda_T}{6\pi} - i \frac{k}{6\pi} \right) \mathbf{1}. \end{aligned} \quad (\text{B9})$$

We now observe that Maxwell's equations are basically a macroscopic theory, so pointlike objects represent some microscopic structure that cannot be resolved on the scale of the wavelength of light. Hence, all the functions relative to physically measurable quantities can be considered to apply only to  $r > a$ , where  $a$  is some microscopic length, while the  $\delta$  function can be replaced by a constant that is the inverse of the volume  $\Delta = \frac{4}{3}\pi a^3$ . This argument allows for an interpretation of the cutoff momenta ( $\Lambda_T, \Lambda_L$ ). In order to obtain such information, we consider the unregularized free-space Green tensor [see Eq. (B3)] and calculate the mean value assumed by this function in a small sphere centered at  $r=0$  and whose radius  $a$  is comparable to the dimensions of the pointlike scatterer. After some algebra, we obtain

$$\begin{aligned} \mathbf{G}^0(r=0, \omega) &= \frac{1}{\Delta} \int_{\Delta} d\mathbf{r} \mathbf{G}^0(r, \omega) \\ &= \left( \frac{1}{4\pi k^2 a^3} - \frac{1}{4\pi a} - i \frac{k}{6\pi} \right) \mathbf{1}. \end{aligned} \quad (\text{B10})$$

Assuming this mean value as the value taken in  $r=0$  by the Green function and comparing Eq. (B9) and Eq. (B10), we obtain

$$\Lambda_L^3 = \frac{3}{2a^3}, \quad \Lambda_T = \frac{3}{2a}. \quad (\text{B11})$$

By this procedure the singularity at  $r=0$  has been removed and a relationship between the cutoff momenta and the dimension of the scattering object has been found.

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